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Research

IGNITION AND COMBUSTION OF
BULK METALS
IN A MICROGRAVITY ENVIRONMENT

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ABSTRACT

Knowledge of the oxidation, ignition and combustion of bulk metals is important for fire safety in the production, management and utilization of liquid and gaseous oxygen for ground based and space applications. This report summarizes research under NASA support to investigate the ignition and combustion characteristics of bulk metals under varying gravity conditions. Metal ignition and combustion have not been studied previously under these conditions and the results are important not only for improved fire safety but also to increase knowledge of basic ignition and combustion mechanisms. The studies completed to date have led to the development of a clean and reproducible ignition source and diagnostic techniques for combustion measurements and have provided normal gravity combustion data on ten different pure metals. Metal specimens were ignited using a xenon short-arc lamp and measurements were made of the radiant energy flux, surface temperature history, spectroscopy of surface and gas products, and surface morphology and chemistry. Elevated gravity was provided by the University of Colorado Geotechnical Centrifuge.

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I. SUMMARY OF THE OBJECTIVE AND VALUE OF THIS RESEARCH

Many aspects of the combustion of metals remain unknown in spite of the practical problems represented by combustion of these materials. The knowledge of the oxidation and combustion of metals is a very important topic for the management of cryogenic fuels or oxidizers in space (1). Industries supplying cryogenics to NASA have displayed an interest in metal combustion because of the possible accidental ignition of tankers carrying liquid or gaseous oxygen. Components of cryogenic pumping and handling systems have experienced accidental ignition which has led to destruction of propulsion systems. Another area of concern is the possible ignition and combustion of metal components of spacecraft structures. Since metals are known to be combustible, it is important to identify flame-inhibiting atmospheres for any space station use of such materials. Identification of ignition mechanisms may also suggest practical procedures for minimizing the possibility of ignition.

This report describes results of a research program under NASA support (NAG 3-1257) to investigate the ignition and combustion characteristics of bulk metals in a microgravity environment. Metal ignition has not been investigated previously under these conditions and the results will be useful in clarifying the ignition and combustion mechanisms by eliminating one of the factors complicating the interpretation of ground-based measurements. The experiments done to date have involved measurements at normal gravity to develop a clean and reproducible ignition source. The ignition source that we have found most effective is a short-arc xenon lamp which has successfully ignited a large number of metal specimens by radiant heating in a pure oxygen environment. A compact combustion chamber with full optical access has also been constructed for use in the experiments. The feasibility of remote operation of the combustion chamber for the proposed aircraft low gravity experiments has been demonstrated by elevated gravity tests in a geotechnical centrifuge. Finally, a number of compact and reliable measurement techniques have been developed and demonstrated in the normal gravity experiments. These measurements provide a database of normal gravity measurements for comparison to elevated gravity and microgravity measurements made with the same apparatus.

II. BULK METAL IGNITION USING A XENON SHORT-ARC LAMP

In this research we have used a 1000 W short-arc xenon lamp (2,3). This light source has several advantages over laser light sources. The size and fragility of lasers makes them difficult to use under high gravity loads in a centrifuge and low gravity flight conditions. Also, a xenon lamp radiates approximately 80% of its power at wavelengths below 1060 nm. This results in a decreased input power requirement for xenon arc lamps over CO₂ lasers because absorptivity of metals increases significantly with decreasing wavelength below 1060 nm. Finally, the price of a complete 1000W xenon arc lamp system is approximately \$5,000, whereas a comparable CO₂ laser would cost approximately \$40,000 to \$50,000.

The metals tested in this investigation, iron (Fe), titanium (Ti), zirconium (Zr), magnesium (Mg), zinc (Zn), tin (Sn), copper (Cu), vanadium (V), manganese (Mn) and aluminum (Al) were selected because of their simple composition (pure metals instead of multi-component alloys to avoid complication in spectroscopic and metallographic studies). These samples were also chosen to study the two different combustion modes experienced by metals: heterogenous or surface oxidation, and homogenous or gas phase reaction. In addition, these specimens exhibit a wide variety of melting and boiling temperatures, heats of combustion, oxide formation processes and adiabatic flame

temperatures. Copper was selected since no reported cases of copper rod burning at normal gravity were found in the literature. The experimental approach provides surface temperature profiles, ignition temperature values, pressure records, spectroscopic records, surface morphology, x-ray spectrometry of metal specimens and their combustion products and high speed cinematography of the heating, ignition and combustion stages of the metal specimen.

The experimental system is shown in Figure 1. The high-intensity, non-coherent light comes out of the lamp in a highly collimated beam which is focused down to a 5 mm spot on the top surface of a metal sample providing approximately a 1 MW/m^2 power density. The cylindrical metal specimen is 5 mm in diameter and 5 mm high and it is held in place by an alumina-silica holder that rests in a ceramic pedestal. These components are located inside a 4.5 liter stainless steel cylindrical chamber which permits input radiation through the top quartz window. Optical access for the camera and spectrograph is provided through three fused-silica side windows. An oxygen environment (99.5% min. O_2) is used in all tests at an absolute pressure of 0.1 MPa. Measurement of surface temperature histories during the heating phase are provided by type B thermocouples, while a solid-state piezoresistive pressure transducer monitors the chamber pressure throughout the run. High-speed cinematography provides a qualitative visual record of the transient process and an estimate of flame propagation rates. A broadband spectrometer and array detector system is used to obtain information on reacting species and products through time and space resolved spectroscopic measurements; the continuum spectrum from the molten material on the reaction interface gives an estimate of color temperature during the ignition and combustion phases. Metallography of specimens before and after combustion is performed in order to identify the reactants and products and their location on the metal sample; surface morphology studies of specimens, quenched at various stages during the process, help to define the thickness and nature of the metal oxide layer formed around the specimen during the heating phase. X-ray spectrometry and chemical analysis of the combustion products complement the metallography study.

Initial testing of the short-arc lamp was performed with simple heating and melting experiments with the different metals in air and atmospheric pressure conditions. Melting was successfully achieved in all cases in times between 15 and 25 seconds. The lamp was then tested for radiant ignition of metal samples in a pure oxygen atmosphere. All these metals exhibit ignition and combustion behavior varying in strength and speed. Film data, temperature profiles, spectroscopic information and metallographic analysis reveal the dynamic evolution of the heating, ignition and combustion phases. These phases are characterized by well-defined surface structure changes, surface temperature, luminosity and chemical activity. Values of ignition temperatures below (as with Fe, Ti and Zr), above (as with Sn, Zn and Mg) or in the range of the metal melting point (as with Cu) are obtained from the temperature records.

A very interesting and unexpected result of this investigation was the ignition and combustion of copper samples by radiant heating. Previous work (4) has classified copper rods as non-flammable even at the highest possible pressure (69.5 MPa in some cases) due to the sudden extinction of the rod after ignition on upward propagation studies at normal gravity using the promoted combustion test (with 3.2 mm diameter rods). Under our experimental configuration, clear evidence of ignition and subsequent downward propagation on copper rods (5 mm in diameter and up to 30 mm long) was obtained at a pure oxygen pressure of 0.1 MPa. X-ray spectrometry of the combustion products provided further proof of complete metal oxidation. The difficulty of burning copper rods in the upward propagation mode at normal gravity may be due to the low heat of combustion of copper in the presence of high heat losses. Under this configuration, the detachment of the first molten ball

from the vertical rod removes the thermal mass that preheats the solid metal in front of it and causes a rapid cooling of the reaction zone with self-extinguishment following afterwards. Since this growth-and-detachment event is absent under microgravity conditions and convective heat transfer losses are significantly reduced, successful upward propagation tests with 3.2 mm diameter copper rods at oxygen pressures as low as 6.9 MPa have been obtained in low-gravity combustion experiments by Steinberg, et al. (5).

In order to conduct elevated gravity experiments, the experimental apparatus will be mounted on the University of Colorado Geotechnical Centrifuge. Experiments similar to those already conducted at normal gravity will be conducted to provide data at 2 to 20 g. The centrifuge was dedicated in 1988 and has received National Science Foundation recognition as a National Centrifuge Laboratory. The centrifuge is equipped with 60 channels for instrumentation transmission, a remotely controlled signal-conditioning system which, with an onboard computer, enables the experimenter to set the gains and offsets of all signal channels from outside the centrifuge during spinning. The amplified analog signals can be relayed by a data-acquisition system to a computer with large disk capacity. The system has proven software with an analog-to-digital conversion capability of one million data points per second. Preliminary results have been obtained in the centrifuge and are discussed below.

III. DIAGNOSTIC MEASUREMENTS

Metal Surface Thermometry

For the internal near surface temperature measurement, a thermocouple well 0.635 mm in diameter is drilled into the specimen from below to within approximately 0.254 mm of the top surface. Type B (Pt-6%Rh/Pt-30%Rh) thermocouples, 0.2mm in diameter, have been found to be the most effective in accurately measuring the near-surface temperature up to the point where the combustion wave reaches the thermocouple junction. Figure 2 shows the temperature histories of some of the metal specimens studied at normal gravity. Figure 3 shows the preliminary results obtained from the centrifuge experiments. The figure gives the temperature vs. time behavior of titanium samples subjected to several gravity loads. Estimates of color temperature of the specimen surface after ignition will be obtained by spectroscopic techniques from the continuum spectrum from the molten material on the reaction interface.

Surface and Flame Visualization

Visualization is required to determine the structure of the combustion process. High-speed cinematography provides a qualitative visual record of this dynamic event and an estimate of flame propagation rates. Movies of the normal-gravity experiments have been obtained with a Teledyne DBM-45, 16 mm high-speed motion picture camera at speeds between 200 to 400 frames per second; this camera is currently on loan from the NASA -Lewis Research Center. The film used has been the Kodak Ektachrome 7240 Color Reversal Film (ASA 125). The DBM-45 camera is a compact, battery-powered unit capable of operating under severe environmental conditions, making it also an ideal candidate for the centrifuge and parabolic flight experiments. Flame structure and convective effects are better visualized with an index-of-refraction technique such as the schlieren method. The advantage of this method is that it responds to index-of-refraction gradients rather than emitted light. Thus in the gas phase, one visualizes the density (or temperature) field. Schlieren is simpler to implement and has less restrictive optical component mounting tolerances than interference methods. The image can be readily recorded on an array detector for computer processing. We are in the process of designing a compact schlieren system that can be used in any gravity environment.

Combustion Spectroscopy

In addition to visible light imaging, spectral information about the products of combustion are useful in determining fundamental combustion-kinetic-mechanism information. Previous studies (6,7) have demonstrated that the optical spectra of the burning surface are a strong function of time and the process details, and can be used to aid in characterizing the process.

To demonstrate the technique we have used an existing uv-visible spectrometer (Princeton Instruments IRY-700 detector mounted on a Jarrell-Ash Monospec 27 Spectrograph) to obtain spectroscopic information on a variety of metals under normal gravity conditions. Figure 4 shows the emission spectra from the ignition and combustion of magnesium samples in oxygen. We have found that such spectra provide essential information on gaseous combustion intermediates and products for our studies. Unfortunately, the present IRY-700 detector is not suitable for remote use at high and low gravity. We therefore propose in future studies to purchase an Alton Instruments Model PR-10C Spectrometer System, which can withstand the stringent operating constraints, improve the spectral resolution by a factor of 3, and increase the data collection rate by factor of 30.

Surface Morphology and Chemical Analysis

The character of the surface has a major effect on the nature of ignition and burning. By analyzing the surface morphology before and after combustion, one may determine the effect of surface structure. On the other hand, the ignition of many metals involves the formation of solid or molten oxides of the metal on the surface of the heated specimen through which oxygen subsequently must diffuse. A more complete understanding of the ignition mechanism for these metals requires monitoring the formation of these oxide layers and their subsequent chemical transformations. In particular, we anticipate that the formation and influence of these oxide layers will be substantially different in a low-gravity environment. The absence of buoyancy-induced free convection will alter the advective transport, thereby changing the chemical environment immediately adjacent to the metal surface. This will alter the chemical nature of the oxide layer and also possibly influence oxygen penetration and subsequent ignition of the underlying metal surface. The growth of this oxide layer can be observed by arresting the oxidation process prior to combustion in order to study its structural and chemical characteristics.

To study these phenomena, we have been using two different instruments to examine metal samples burned under 1-g conditions. Morphological analysis is obtained with a Cambridge Stereoscan 250-Mk3 scanning electron microscope (SEM) which is equipped with digital image enhancement to facilitate ultra-high resolution of minute structural features. Quantitative analysis of the chemical composition of the sample has been performed with a JEOL JXA-8600 electron microprobe (EMPA) with combined wavelength dispersive spectroscopy (WDS) and energy dispersive spectroscopy (EDS). The Center for Combustion Research employs a full-time, college-trained electron microscopist to maintain and operate our SEM and electron microprobe facility and to assist us in developing special techniques to study our combustion samples. Figure 5 shows an example of the use of the above techniques in determining the combustion mechanism of copper. The SEM photograph shows a 6000X magnification of the top outside layer of a copper specimen quenched during combustion in an oxygen environment at 0.1 MPa. The same equipment and analytical techniques will be applied for unburned, partially-oxidized and burned metal samples under different g-level conditions.

IV. MODELING OF THE IGNITION AND COMBUSTION PROCESS

We have begun numerical modeling studies of the heating, ignition and combustion of the metal

specimens studied experimentally. The modeling includes all relevant heat transfer mechanisms to the metal specimen located on the ceramic base and natural convection in the gas phase surrounding the specimen. The calculations completed to date have considered the input radiation due to the xenon short-arc lamp, and the subsequent heating of the specimen to the ignition temperature (3). The modeling calculations have considered the effect of gravity level and oxygen pressure on the heating of the metal. Comparison of the calculations with measured temperature rise in the metal gave excellent agreement. It was found that significant differences in the time history of the metal temperature were predicted for different gravity levels. The calculations also showed that the heat loss by natural convection was important and varied substantially with gravity level.

V. INDUSTRIAL AFFILIATIONS

Industrial affiliations and interaction have been extensive in the progress of this research. We have participated in the bi-annual conference on Flammability and Sensitivity of Materials in Oxygen Enriched Atmospheres sponsored by the American Society of Testing and Materials (ASTM). This meeting is a forum for information exchange among those organizations concerned with safety questions related to handling liquid and gaseous oxygen. The ASTM is also responsible for developing tests to evaluate and compare the risks associated with metals used in oxygen environments. Among the attendees at these meetings are equipment manufacturers, process companies, and research organizations. As one direct result of our participation in these meetings, we are interacting with PRAXAIR, Inc. on problems that they have experienced with ignition of liquid oxygen distillation columns made of aluminum.

The Lockheed Engineering and Sciences Company, in conjunction with the NASA White Sands Test Facility has been conducting tests of assisted ignition of metal rods in normal and microgravity (5). Dr. Theodore Steinberg from that group has served as a Research Affiliate to our activities on metal ignition.

Finally, we have a good working relationship with the NASA Lewis Research Laboratory and are currently using one of their cameras for high speed cinematography of the ignition and combustion of our metal specimens. This is a part of our overall goal of testing and validating all diagnostics to be used in the microgravity experiments beforehand in normal gravity experiments.

VI. FUTURE RESEARCH

Future research will use the apparatus and techniques developed here to complete the elevated gravity measurements, to conduct the microgravity measurements, and to develop the modeling of the overall ignition and combustion process. The normal gravity experiments have provided new spectroscopic and electron microscopic data on a number of pure metals. The elevated gravity measurements are to be completed in the Geotechnical Centrifuge operated by the University of Colorado. Metal combustion has already been achieved and monitored in the centrifuge for a small number of specimens. Since the time required for heating, ignition and combustion of the metal specimens is typically 20-25 sec, the microgravity environment will be provided by a DC-9 aircraft flying Keplerian trajectories operated by NASA/Lewis Research Center. The modeling studies will focus on the description of the essential effects of gravity body forces on the ignition and combustion process and the details of the ignition mechanism.

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IX. PERSONNEL AFFILIATED WITH THIS RESEARCH

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2. John W. Daily, Professor of Mechanical Engineering, Co-Principal Investigator.
3. Angel Abbud-Madrid, Graduate Research Assistant, Ph.D. candidate.
4. Thomas J. Feiereisen, Graduate Research Assistant, M.E. Degree, December 1993.
5. Gregory J. Fiechtner, Post-Doctoral Fellow.
6. Matthew Fuller, Undergraduate student.

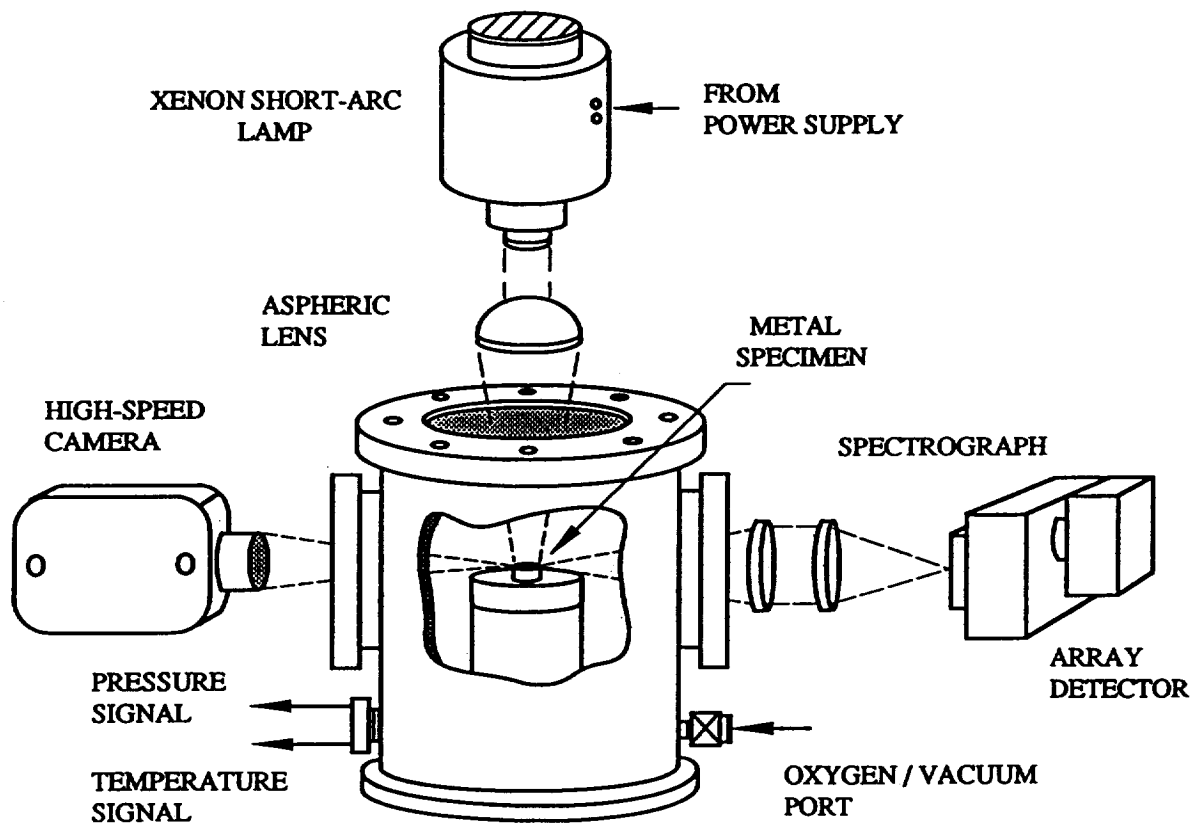


Figure 1. Experimental system used in the study of ignition and combustion of bulk metals under elevated, normal and reduced gravity conditions.

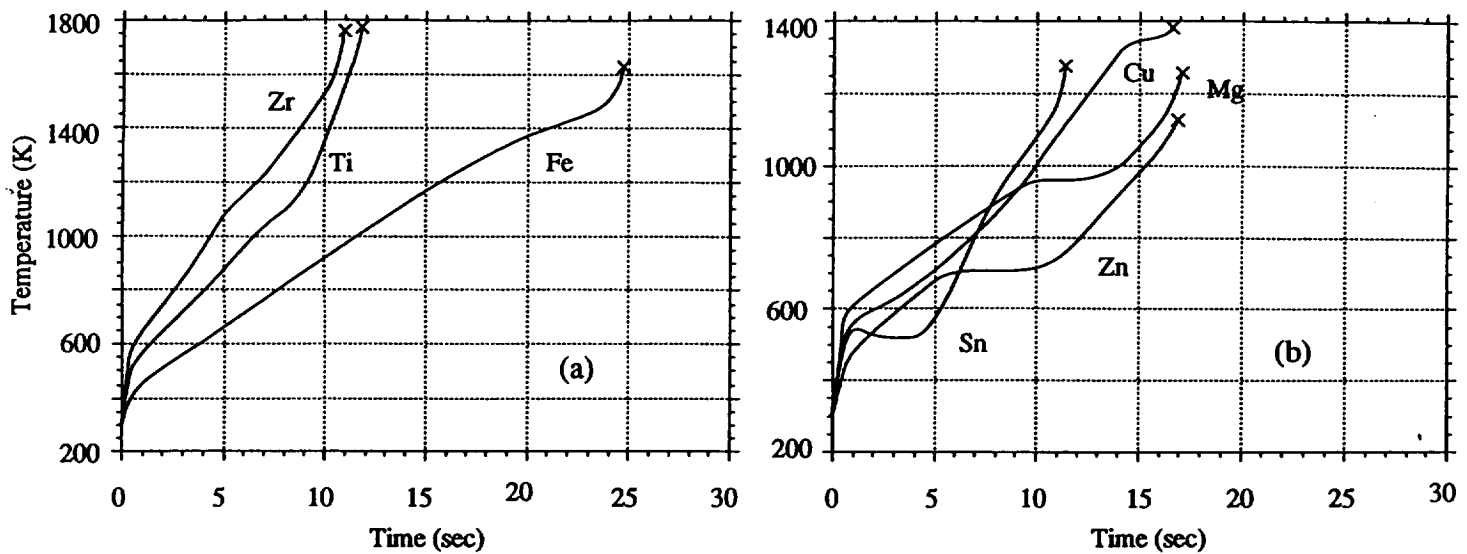


Figure 2. Temperature histories of various pure bulk metals in normal gravity at 1 atm . The ignition temperature is identified as the point of inflection after which the slope increases rapidly until the thermocouple breakpoint. For Fe, Ti and Zr in Fig. 2a the ignition point occurs below the melting point of the metal. The temperature profiles for Sn, Zn, Mg and Cu (Fig. 2b) show an intermediate plateau which corresponds to the solid-liquid phase transition. For these metals their ignition temperature lies above their melting point.

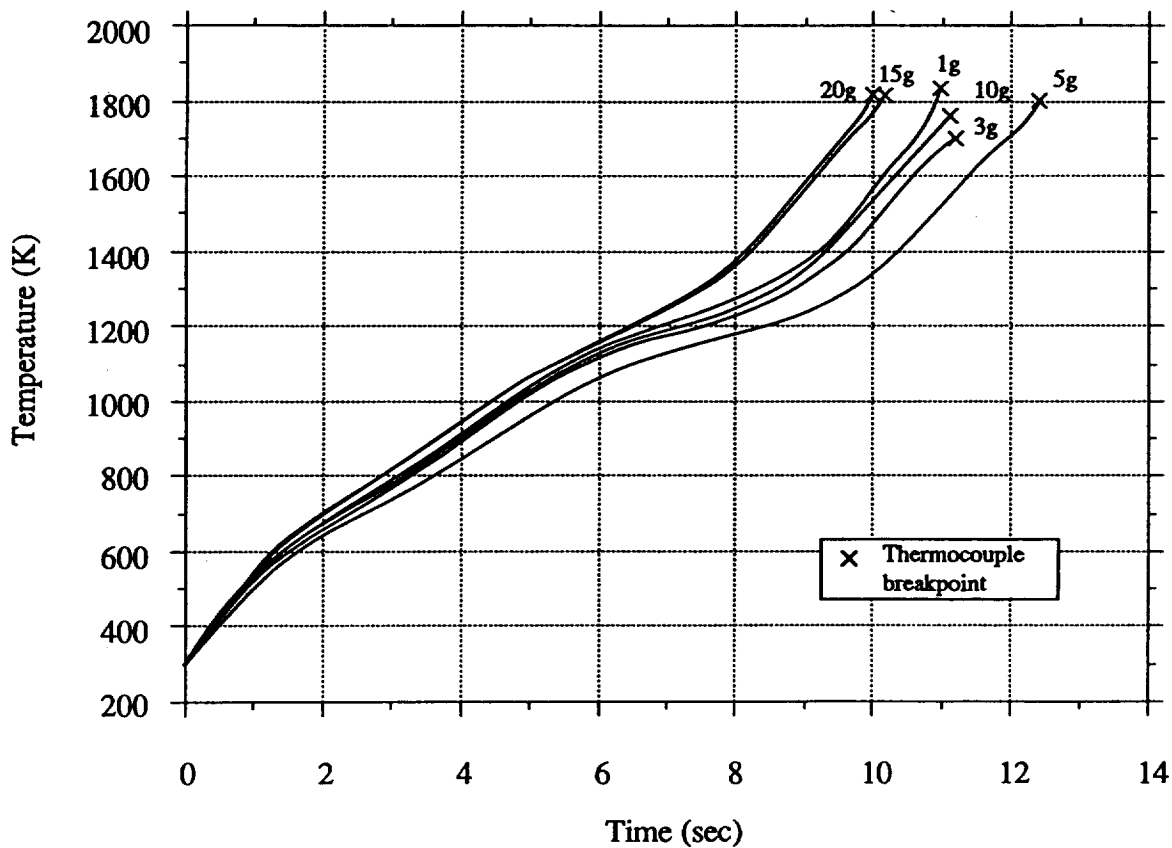


Figure 3. Temperature vs. time behavior of Ti samples subjected to different gravity loads in a geotechnical centrifuge. As the gravity level is increased from 1g, so is the ignition delay time. This trend is seen to reverse after the 5g curve, leading to faster ignition times for metal specimens under higher gravity loads.

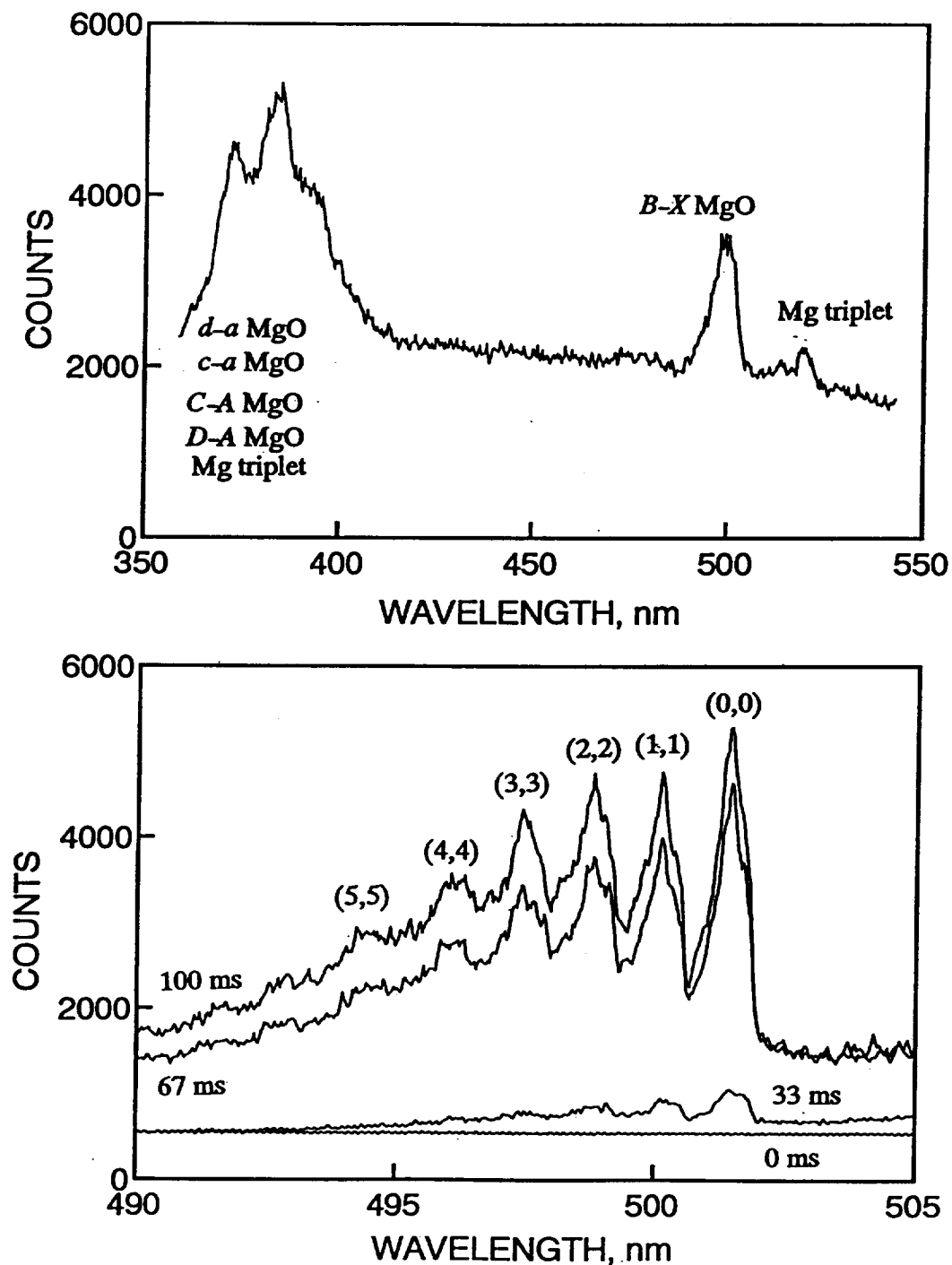


Figure 4. Emission spectra taken during the ignition and combustion of magnesium samples in oxygen. All spectra are taken with a 33- μ s integration time and no signal averaging. The top curve was taken approximately 100 ms after ignition using a 300 groove/mm grating, and includes many spectral features of both MgO and Mg vapor. The bottom curves were taken in 33-ms intervals using a 2400 groove/mm grating to highlight the vibrational features of the green *B-X* system of MgO. Emission spectra from other metal-oxygen reactions have also been recorded, including titanium and zinc.

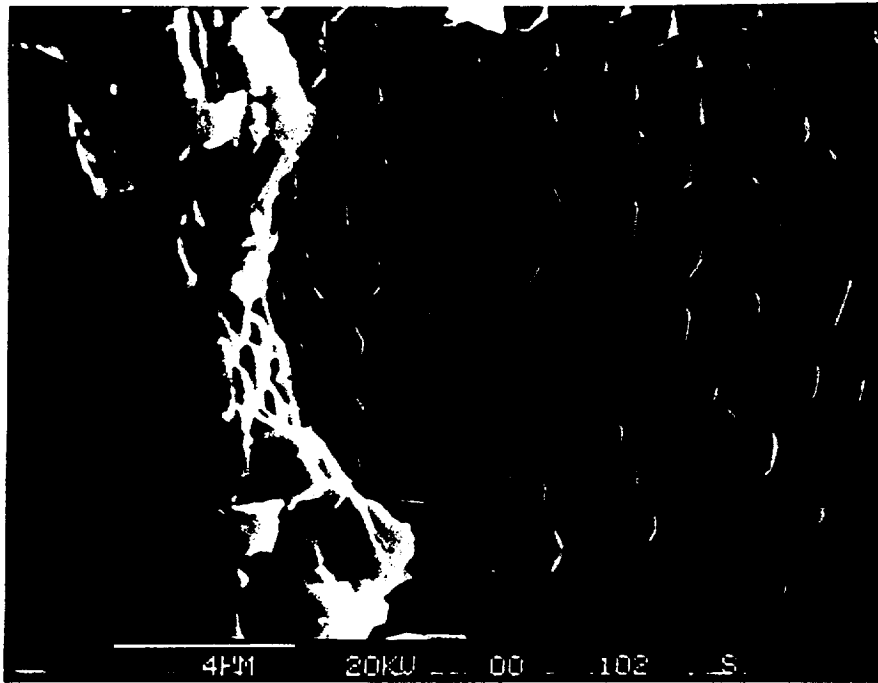


Figure 5. Scanning electron microscope photograph (6000X magnification) of the top outside layer of a copper specimen quenched during combustion in a pure oxygen environment at 0.1 MPa. Copper crystals belonging to the hexoctahedral crystal class appear to have been deposited on a Cu+O matrix by the condensation of copper vapor after the quenching event.